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FUEL-RICH SOLID PROPELLANT BORON COMBUSTION

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Annual Report Covering
4/1/81 - 3/31/82

Air Force Office of Scientific Research/NA
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A thorough critical assessment of the state-of-the-art of boron fuel ignition/combustion has been carried out and areas of deficiency defined. An improved boron single particle ignition model treating the effects of water vapor on particle ignition has been developed. Boron single particle combustion models which include the effects of finite kinetics and correctly predict transition from a second-order dependence of burn time on particle size to a first-order dependence for small particle sizes have also been cont

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20. ABSTRACT (continued)

cont developed. Considerable progress has been made in developing an experimental apparatus and diagnostics for studying single boron particle ignition and combustion processes for small (less than 25 micron diameter) particles and for studying the kinetics of the critical (for ignition) liquid boric oxide — water vapor reaction. In addition, a test bed for study of the processes involved in burning of compacted boron solid fuel grains in a hot air crossflow has been designed and is under construction. Finally, a simplified model of processes occurring in a perfectly stirred reactor with a cloud of boron particles as the fuel has been developed and is being used in design of a test apparatus for study of boron dust cloud ignition and combustion.

I. RESEARCH OBJECTIVES

(1) Modify the existing analytical model of boron ignition by King to include treatment of finite-rate kinetics for the reaction of water gas with surface oxide.

(2) Develop a mechanistically accurate model for combustion of clean boron particles, with allowance for finite rate kinetic steps and multistage oxidation as necessary.

(3) Upgrade an existing boron cloud ignition model to include a more detailed submodel of single particle ignition phenomena and to permit prediction of ignition delay times as well as critical conditions required for cloud ignition.

(4) Modify the aforementioned single particle boron ignition model to treat effects of ignition modifiers.

(5) Develop stirred reactor and directed-flow reactor models of combustion of boron dust clouds utilizing the unit ignition and combustion models developed in (1), (2) and (4).

(6) Evaluate the feasibility of various approaches to determining the kinetics of the reactions of condensed-phase boron with various gaseous oxidizers and, if feasible, perform detailed design of such an experiment.

(7) Critically analyze the literature regarding condensation of B_xO_y and HBO_2 gases produced by the combustion of boron particles and define experiment(s) to quantitate this phenomenon.

(8) Experimentally evaluate the kinetics of $B_2O_3(l) + H_2O(g)$ using a flat-flame burner procedure.

(9) Experimentally define the intermediates appearing in the combustion of boron.

(10) Obtain ignition and burning time data for single boron particles in the 5 to 25 micron diameter size range using a flat-flame burner procedure.

(11) Study the conversion of boron in a center toroidal recirculation zone reactor as a function of several independent variables.

(12) Experimentally investigate the flame structure associated with a consolidated boron grain burning in an air crossflow.

(13) Experimentally evaluate the effects of such potential boron ignition promoters as magnesium, fluorine, and lithium fluoride on the ignition characteristics of boron particles.

(14) Measure ignition delay times of boron dust clouds and critical conditions required for ignition of such clouds as a function of the important independent parameters.

(15) Measure the flame propagation rates of boron dust clouds as a function of the important independent parameters.

II. ACCOMPLISHMENTS AND PROGRESS DURING THE PAST YEAR (4/1/81 - 3/31/82)

Significant progress was made on several of the tasks listed in the previous section during the past year. First, a thorough critical review of domestic and foreign literature in the area of boron ignition and combustion was carried out. Experimental and theoretical studies of both single particle and boron dust cloud ignition/combustion behavior were included. Attempts were made to reconcile differences between investigators where possible. In addition, the relevance of the work reviewed to practical combustor situations was delineated. From all of this, gaps in existing knowledge were defined and factored into this program so that the current investigation might aid in closure of these gaps.

As part of the aforementioned review, shortcomings of existing boron single particle ignition models and approaches to improving them were defined. During the past year, a new model of boron ignition in wet and dry atmospheres, involving major modifications to the boron ignition model developed by King in the early 1970's, was developed. As before, the model centers around time-dependent mass balances on boron and its oxide coating (with geometrical relationships converting these to time-dependent differential equations for particle radius and oxide thickness) and a particle energy balance (in the form of a differential equation for temperature or fraction of boron melted, depending on whether the particle is at its melting point). These equations relate the time rate of change of particle radius, oxide thickness, and particle temperature (or fraction melted) to various source/sink terms associated with energy exchange with the surroundings, reaction of oxygen with boron at the boron/boron oxide interface, evaporation of boric oxide, reaction of boric oxide at the outer edge of the oxide coating with ambient H_2O to form $HBO_2/H_3B_3O_6$, and possibly reaction of water at the B - B_2O_3 interface with boron to form B_2O_3 and H_2 (which then reacts with oxygen, as available, at the outer edge of the oxide layer) or with boron and B_2O_3 to form $H_3B_3O_3$. (Several scenarios are examined as regards the effects of H_2O on boron particle ignition — it is not yet clear which is most realistic.) Expressions are developed for each of these source/sink terms and the differential equations are then integrated simultaneously for a given set of boundary (ambient) conditions and initial conditions until it becomes obvious that thermal runaway will not occur (no ignition) or until such runaway does occur (thus yielding the ignition time). Several major changes have been made in the current analysis relative to the earlier work by King:

(1) In King's past work (and many of the other past boron ignition models) it was assumed that the entire resistance to reaction of O_2 with boron in the presence of an oxide coating was associated with dissolution of O_2 in the liquid oxide and transport across it to the B - B_2O_3 interface. Now series resistances for kinetic rate limitations on the interface reaction, dissolution and transport of O_2 across the liquid oxide layer and diffusion of gaseous O_2 from the free-stream to the oxide coating outer surface are included.

(2) The expression used to represent the resistance for transport of dissolved O_2 across the liquid oxide layer has been modified in light of additional data and first principles calculations.

(3) Blowing effects on mass and energy transport in the gas surroundings are now included.

(4) A momentum equation has been added for calculation of particle slip in a moving gas stream and the effects of this slip on mass and heat transfer between the particle and surrounding gas are included.

(5) Previously, the reaction of B_2O_3 with H_2O at the outer edge of the oxide layer was treated as being diffusion-limited, leading to overprediction of the effects of H_2O on ignition. Finite kinetics, based on Russian data, are now included in this part of the analysis. Two possible scenarios are treated: (A) surface reaction; (B) volumetric reaction throughout the oxide layer.

(6) Capability of handling possible reactions of H_2O at the B - B_2O_3 interface following diffusion of H_2O across the oxide layer is included. Three possible scenarios are included: (A) No such reaction, (B) $H_2O + 4/3\ B + 1/3\ B_2O_3 \rightarrow 2/3\ H_3B_3O_3$; (C) $H_2O + 2/3\ B \rightarrow 1/3\ B_2O_3 + H_2$ followed by fast reaction of the H_2 so produced with O_2 at the oxide outer surface (depending on the O_2 availability).

(7) A much higher heat of melting of boron, reported in the most recent JANNAF Thermochemistry Tables, is used.

(8) A redefinition of necessary/sufficient conditions for boron ignition is incorporated. In King's previous model, boron melting was always complete prior to final disappearance of the oxide coating: as a result, since the only resistance to reaction of the boron particle with oxygen was proportional to the instantaneous oxide thickness, disappearance of the oxide and thermal runaway coincide. In the new model, however, under some conditions, the oxide removal is completed before the particle has melted. In such cases, a non-negligible period of time is subsequently required to finish raising the particle temperature to the melting point and carrying out the fusion: after this is complete, thermal runaway occurs very quickly. Thus, both completion of oxide removal and particle melting are required for ignition.

(9) As a result of (8), the analysis must be modified to continue in the absence of an oxide layer. Currently, approximate expressions for the rate terms based on a very simplified combustion model are utilized for this stage of the analysis — eventually these will be replaced with equations based on a more sophisticated boron combustion model being developed in parallel with the ignition modeling.

Equation development and programming are complete, and testing of the model against the rather limited Macek data base has begun. More data are required before full testing and calibrating of the model (including selection of the correct scenario where multiple scenarios are included as options) can be completed. Efforts are currently under way to develop the required data base. A particularly interesting preliminary result as regards possible tailoring of particle surroundings conditions in a combustor has been obtained. It is predicted that ignition delay time can be minimized by maintaining the particle in a region where it sees water vapor but no oxygen until oxide removal is

complete and then exposing it to maximum possible oxygen level through thermal runaway (as opposed to keeping the particle in a high O_2 environment throughout the heat-up/ignition process).

Inasmuch as the boiling point of boron is in excess of 3900°K, it is apparent that clean boron particle combustion (subsequent to removal of the oxide coating during the ignition stage) must involve heterogeneous surface reaction processes. As discussed below, however, it is not required that full oxidation to final products occur at the surface. Observations of Macek indicate that for large boron particles, combustion is diffusion-limited while for smaller particles there is a shift towards kinetics control. Moreover, in the case of hydrogen-containing environments not only O_2 but H_2O serve as oxidants in the overall combustion process. Over the past year, a series of models of boron particle combustion of increasing complexity and completeness has been developed for boron single particle combustion in wet and dry atmospheres under diffusion-limited and partially-kinetics-limited conditions.

First, several models were developed for the diffusion-limited combustion of boron particles (valid only for large particles) in dry atmospheres. These models included:

I. Single One-Stage Reaction of Boron to B_2O_3 Gas at the Particle Surface.

II. Reaction of B_2O_3 Gas at the Particle Surface to Yield B_2O_2 Gas, with Subsequent Diffusion-Limited Conversion of B_2O_2 + O_2 to B_2O_3 in the Gas Phase.

III. Reaction of B_2O_3 Gas at the Particle Surface to Yield BO Gas, with Subsequent Diffusion-Limited Conversion of BO + O_2 to B_2O_3 in the Gas Phase.

IV. Reaction of B_2O_3 Gas at the Particle Surface to Yield a BO/B_2O_2 Gas Mixture (Relative Amounts Determined by Equilibrium Considerations at the Surface Temperature) at the Particle Surface, with Subsequent Diffusion-Limited Oxidation to B_2O_3 in the Gas Phase.

V. Equilibrium Throughout the Gas Phase (Shifting Due to Temperature Variations) with Equilibrium Between $B(\ell)$ and the Various Gas Species at the Surface. B, BO, B_2O_2, BO_2, B_2O_3, O_2, N_2 Species Included.

The fifth model is, of course, the most accurate representation of diffusion-limited combustion of a boron particle in a dry atmosphere. It is interesting to note, however, that over a wide range of conditions Models II-V yield nearly equal predictions for burn rate (though II-IV yield serious errors in surface temperature, rendering them unsatisfactory for prediction of ambient temperature-ambient oxidizer concentration extinguishment boundaries, extinguishment being postulated to occur when the partial pressure of B_2O_3 at the surface exceeds the vapor pressure of B_2O_3 at the surface temperature.) Moreover, it is found that the burn rate under diffusion-limited conditions in dry atmospheres can be calculated quite well using:

$$\dot{m} = \frac{\rho D}{r_s} \ln (1 + B) \qquad (1)$$

$$B = 0.677\, X_{O_2,\infty} = \frac{2MW_B}{MW_{O_2}} X_{O_2,\infty} \qquad (2)$$

where $\dot{m}$ is the mass burning flux, ρD is the gas density-diffusivity product, r_s is the particle radius, and $X_{O_2,\infty}$ is the ambient oxygen mass fraction. These equations are found to give good agreement with Macek large particle burn-time data for reasonable values of transport parameters, while Model V yields extinguishment boundary predictions consistent with Macek's semi-quantitative experimental observations.

Next three models (still limited to dry atmospheres) with allowance for finite rate kinetics for the surface reactions were developed. In the first very simple model (found to be inadequate) Model I was modified to incorporate finite kinetics for the single reaction of boron plus oxygen to B_2O_3 at the surface. In the second model, finite rate reaction of B_2O_3 gas with boron at the surface to yield B_2O_2 with subsequent diffusion-limited gas-phase reaction of B_2O_2 with oxygen in the gas-phase to produce B_2O_3 was considered. (For sufficiently small particles, breakthrough of oxygen to the surface occurs with this model and finite rate reaction of O_2 with boron to yield B_2O_2 at the surface is then added in.) Finally, Model V was modified to treat finite-rate kinetics for reaction of all species with a molar B/O ratio less than 1.0 with condensed phase boron at the surface, with shifting equilibrium prevailing in the gas-phase.

For low values of activation energy for the surface reactions (realistic), the second and third models were found to yield nearly equal predicted burn rates over the entire particle size regime studied (1 to 200 micron radius) with a gradual shift from diffusion control to kinetics control with decreasing particle size. (As in the diffusion-limited case, the simpler model yields unrealistically high surface temperatures, however.) With reasonable values for the surface reaction activation energies, both the Macek small particle size data and large size data are fit quite well by these models.

Recently the more complete models (Model V and its counterpart with kinetics limitations included) have been extended to treat wet (B-H-O-N) systems. Preliminary comparison with very limited Macek data indicates reasonable agreement of model and data. In the diffusion-limited case, Eqn. 1 is again found to represent the results of the detailed calculations quite well, with B now being equal to

$$0.677\, X_{O_2,\infty} + 0.602\, X_{H_2O\infty} = (2MW_B/MW_{O_2})\, X_{O_2,\infty} + (MW_B/MW_{H_2O})\, X_{H_2O\infty} .$$

Further work currently in progress includes:

(1) Inclusion of radiative heat losses (currently neglected).

(2) Treatment of effects of B_2O_3 condensation away from the particle (expected to have second-order effects).

(3) Allowance for $dT_p/dt \neq 0$ in the energy balance to permit more accurate treatment of the period between oxide removal and completion of boron particle melting in the previously described ignition model.

(4) Extension to inclusion of carbon species (B-H-O-N-C system).

(5) Allowance for finite-rate limitations on key gas-phase reactions.

In another task, the feasibility of studying the kinetics of oxidation of solid boron with oxygen in the 2000-2400°K region (to provide needed information for the ignition and combustion models) in an apparatus where an oxygen-containing gas stream either flows through a nozzle throat made of boron or impinges supersonically on a boron target has been examined. It has been concluded that providing sufficiently high mass transfer coefficients to prevent the kinetics resistance from being overshadowed (and masked) by diffusional resistance is essentially impossible, given the expected approximate kinetics of the reaction. Accordingly, it is recommended that this approach be abandoned. Alternative approaches to obtaining these kinetic data will be identified and examined for feasibility.

During the first year's experimental efforts, a Combustion Diagnostics Laboratory and a Particle Characterization Laboratory for support of the task to expand the small particle boron ignition/combustion time data base have been established. Precise, fine-cut particle size groups of crystalline and spherodized boron have been obtained through elaborate sizing procedures and characterized. In addition, major effort has been devoted to establishing suitable means for injection of these particles into the products of a flat-flame burner. A precisely controlled flat-flame burner system including a McKenna burner, Teledyne-Hastings gas control system, the aforementioned "home-made" particle feed system, and a motor-driven position controller for moving the burner into and out of the diagnostics field of view has been constructed.

The major diagnostic tools applied to the investigation, thus far, include chopped-scan single frame photography, laser doppler velocimetry, high speed cinematography, and electro-optic scanning of the flow field utilizing a linear photo-diode array (Reticon camera). Gas temperatures are inferred from equilibrium flame temperature calculations based on equivalence ratio and mass fraction of an inert gas diluent. Future efforts will be devoted to establishing flame temperature and particle gas cloud temperature measurements from the Raman shifts induced by pumping optical transitions with a tunable dye laser.

The chopped-scan photography is capable of providing an assessment of ignition times and combustion times of individual particles by passing a rotating vane in front of a view camera lens at a precisely measured rate, and counting events from initial onset of radiation to final burn-out of the particle. Current efforts are being devoted to the design of an appropriate spectral technique for determining the real time evolution of B_2O_3 films on the boron particle surface, which will then provide additional empirical data to refine ignition/combustion modelling.

Laser velocimetry can be used to establish the injection velocity of the carrier gas, the initial velocity of the particle upon entry to the flat-flame burner zone, and the flame velocity in the combustion zone. Each of these measurements is accomplished with the dual-beam fringe type anemometry. For measurements of particle velocity in the combustion zone (where the particle is undergoing constant size reduction), a better approach is laser transit anemometry, or more commonly called "two-spot" anemometry. The dual beam measurements have been made with some degree of success, but the two-spot measurements are being deferred until more data on burn times are obtained.

High speed motion picture films of single particle events have been obtained and while not of high quality, have served to verify some of the conclusions reached in analyzing the chopped-frame photography. A significant draw-back to the technique is that currently available film speeds are too slow to obtain good images at the very high framing rates required to give good fine resolution. Thus, while the films provide excellent qualitative data regarding single versus multiple particle events, they are somewhat deficient as regards quantitative data.

Our most recent efforts have been devoted to making measurements of velocity, size and particle trajectory in the combustion zone using a photodiode array camera manufactured by EG and G Reticon Division. This camera has the advantage of being interfaced to a data acquisition system. In this mode, data can be collected in real time, stored in memory and then processed at a later time, giving a precise history of the particle trajectory, apparent size and radiation intensity.

Each of the foregoing diagnostic systems have been exercised in a single collection mode, but to date no integration or simultaneously accomplished data collection efforts have been achieved. Current efforts are being devoted to development of such an operational mode.

Another task which has been initiated involves characterization of boron dust cloud ignition and combustion in a perfectly stirred reactor. This is essential for proper analytical treatment of stirred reactor regions in a zonal approach to combustor modeling, and for sizing recirculation zones (in terms of residence time) required in a combustor for adequate flame holding. The uniqueness of the problem studied here stems from the superposition of the restrictive ignition and combustion characteristics of an injected boron dust cloud with a given particle size distribution on perfectly stirred reactor dynamics. The general approach to this task is to experimentally evaluate bulk reactor kinetics and flame instability.

A simplified model of a perfectly stirred reactor, including boron particle ignition and ballistics, was developed during this period to guide experiment design and data interpretation. The model predicts thermal and compositional conditions necessary for stable ignition and burning for a given boron particle size distribution and given perfectly stirred reactor geometry and flow conditions. A simplified reaction is assumed for boron burning in air. Three equations for the oxygen/boron mole fraction are derived based on conservation of mass, energy, and available O_2 concentration. Boron particle dynamics are included by combining King's single particle ignition model with an empirical correlation of particle burn times (based on Macek data) evaluated for a perfectly stirred reactor. The three equations for (O_2/B) fraction are solved

simultaneously during the combustion process of a given particle distribution to converge upon a stable stirred reactor temperature and (O_2) volume fraction. Multiple schemes are available for experimentally characterizing the particle size distribution. A Microtrac (SPA) particle size analyzer may be used as a pre-test measurement. A Malvern laser scattering system may be used to obtain real-time particle size distributions. The model is currently being checked out. When checkout is completed, it will be exercised to evaluate the first-order sensitivity of reactor combustion stability to particle size distribution, fuel flow rate, reactor geometry, and global flow conditions. Based upon the results of this evaluation, design of the laboratory stirred reactor will be undertaken. Emphasis will be placed on providing sufficient design flexibility where it is most critical for adequate evaluation of controlling mechanisms.

A final task which has been addressed during the past year is the study of the burning mechanism(s) of compacted boron solid propellant grains exposed to a crossflow of high-temperature air (most germane to the boron solid-fuel ramjet, BSFRJ, concept). A special reaction chamber has been designed for study of the structure and governing mechanisms of a flame established within the turbulent boundary layer of such an ablating and burning boron grain. The chamber is comprised of a series of bolt-together test sections to allow maximum experiment flexibility and is one component of a multipurpose laboratory combustion facility which is currently being assembled. (The laboratory stirred reactor to be designed for the previous task will also be used in conjunction with this facility.) Test sections comprising the reaction chamber include a high temperature air inlet and grain ignition section, common to all test hardware used in the Laboratory Combustion Facility; this is followed by a section for transition to square cross section; flow straightener and turbulence generator sections precede the primary windowed-grain section which is followed by combustion mixing and nozzle sections. Instrumentation ports are provided throughout. The primary windowed grain section is designed to accept either 2-D facing boron grain slabs or, alternatively, to accept a single boron grain slab with an opposed optical access for a controllable external radiation source. Optical access is also provided through the side of the reaction chamber to allow use of both simple and advanced non-intrusive diagnostics being developed in the flat-flange burner study discussed earlier. This experimental hardware has been ordered. Final experiment design and diagnostics selection are under evaluation. A simplified grain ablation model is also being formulated to assist experiment design and data interpretation.

III. PUBLICATIONS

1. King, M.K., "Ignition and Combustion of Boron Particles and Clouds," to be published as invited paper in Journal of Spacecraft and Rockets, mid-1982. Earlier version presented at 1981 JANAAF Combustion Meeting, CPIA Publ. 347, Vol. III, p. 225.

2. King, M.K., "Boron Particle Ignition and Combustion," to be presented as invited paper at 1982 Eastern States Combustion Institute Meeting, Atlantic City, N.J., Dec., 1982.

3. King, M.K., "Single Particle Boron Ignition Modeling," planned for 1982 JANNAF Combustion Meeting, Oct., 1982.

4. King, M.K., "Modeling of Single Particle Boron Combustion," planned for 1982 JANNAF Combustion Meeting, Oct., 1982.

IV. PROFESSIONAL PERSONNEL

Dr. Merrill K. King

Dr. James Komar

Mr. Ronald S. [illegible]

V. INTERACTIONS (COUPLING ACTIVITIES)

Atlantic Research has several advanced development contracts involving use of boron as a fuel. These include ducted rocket, slurry ramjet, and boron solid-fueled ramjet (BSFRJ) programs, funded by AFRPL, AFWAL, DARPA, and NWC. Dr. King, Dr. Komar, and Mr. Fry are all active in these programs, providing modeling and diagnostic support in the areas of boron particle ignition and combustion. As a specific example, Dr. King's ignition model is being incorporated in an analysis being used for design of a boron slurry ramjet combustor. In addition, output from the compacted boron solid fuel program for development of high energy advanced air-breathing propulsion system fuels.

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